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Environmental Sciences Europe

DOI:
10.1186/s12302-020-00367-w

Published: 01/07/2020

Publisher's PDF, also known as Version of record

Environmental Sciences Europe

COMMENTARY

Open Access

Critical accumulation of fertilizer-derived uranium in Icelandic grassland Andosol

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Abstract

Long-term phosphorus (P) fertilizer application can lead to an accumulation of uranium (U) in agricultural soil, potentially posing risks on the environment and human health. In this study, we found that such risks could be severe in two long-term grasslands (Andosol) in Iceland (Sámstaðir and Geitasandur) after about 50 years of P fertilization. At Sámstaðir, where P fertilizers were applied at an annual rate of 39.3 kg ha^{-1} $year^{-1}$, the soil U concentration increased from 0.65 mg kg^{-1} in the unfertilized soil to 6.9 mg kg^{-1} in the fertilized surface soil (0–5 cm). At Geitasandur with P fertilization rate at 78.6 kg ha^{-1} $year^{-1}$, the soil U concentration reached 15 mg kg^{-1}. The average annual U accumulation rates were 130 and 310 µg kg^{-1} $year^{-1}$, respectively. These values were larger, by up to a factor of ten, than any previously reported rates of fertilizer-derived U accumulation. However, the U concentration in one of the applied P fertilizers was 95 mg U kg^{-1} fertilizer, similar to the median value of those reported in previous studies, and thus unlikely to be the only factor leading to the high U accumulation rates. By contrast, as our Andosols had low bulk density within a range of 0.2 to 0.5 g cm^{-3}, the annual U inputs to the 0–5 cm soil were 19 g ha^{-1} $year^{-1}$ and 32 g ha^{-1} $year^{-1}$ at the two sites, respectively, within the range of to-date reported values in agricultural systems. In addition, we found that U was mostly retained in the surface soil rather than mobilizing to deeper soil. This was likely due to the fact that the Andosols were rich in organic matter which promoted U retention. Therefore, the observed high U accumulation rates were a result of the combination of (i) the large amounts of the applied P fertilizers and (ii) the soil properties of the Andosols with low bulk density and elevated organic matter content concentrating U in the upper surface soil. Our study shows that agricultural production systems on Andosols may have already suffered from severe U contamination due to P fertilization. We are therefore calling for future checks and regulations on P fertilizer-related soil U accumulation in these and certain comparable agroecosystems.

Keywords: Uranium, Phosphorus fertilizer, Grassland soils, Andosol, Iceland

Background

Phosphorus (P) fertilizers are primarily derived from phosphate rocks, which, however, contain various levels of uranium (U) [1–3]. The majority (80–90%) of U is transferred to the final fertilizer products during mineral processing [4]. Therefore, U can accumulate in agricultural soil following prolonged P fertilizer application [5–9]. After this risk being first mentioned by Rothbaum et al. [10], the consensus has been reached that P fertilizer-derived U accumulates in the topsoil of agricultural fields [5–11]. Chemical toxicity of U is of greater concern than its radiotoxicity, due to the low intrinsic specific radioactivity of ^{238}U [12]. The most sensitive adverse effect of U on human being is chemically induced toxicity to the kidney via food and water intake [13]. Since the transfer factor of U from soil to plant is below 1% [8, 14], the U uptake by plants and then entering the food chain is not a predominant health issue [13]. However, it has been suggested that drinking water can become a main source of human U intake [15, 16]. A number of studies indicate the transfer of fertilizer-derived U to water bodies [16–19].

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Despite potential negative impacts of U on human being and the environment, there still is a lack of regulations on the limitation of U in P fertilizers both at the regional and global scales. Worldwide, Canada, rich in U resources and with a long history of U exploration, mining and generation of nuclear power [20], is the only country that has implemented a soil quality guideline of 23 mg U kg^{-1} soil for agricultural land use to protect the human and environmental health [21]. However, internationally there still are no limitations for U content in fertilizers [22]. Yet, an increasing number of studies report an accumulation of fertilizer-derived U in agricultural soils or in groundwater [16, 19, 23]. The reported accumulation rates of fertilizer-derived U in soil are in the range of 0–130 μg kg^{-1} $year^{-1}$ with median value of 7.65 μg kg^{-1} $year^{-1}$ [24]. Uranium concentrations in soil have been reported to range from 0.3 to 11.7 mg kg^{-1} with an average background concentration of 2 mg kg^{-1} [21]. Therefore, the fertilizer-derived U accumulation can become a cumulative issue after hundreds of years of mineral P fertilization. Many studies have confirmed that fertilizer-derived U will increase soil U contents, though only marginally and not necessarily to a degree that it significantly increases U exposure to human being via food or drinks [24]. As a result, current pressure on governmental legislations is low to set up a guideline value for U in fertilizers.

Volcanic soils (Andosols), covering approximately 124 million hectares of the land surface, are rich in organic matter and mineral nutrients, have high water-holding capacity, and thus are considered to be important agricultural soil resources in, e.g., Japan, Iceland, and New Zealand, as well as in several tropical areas [25]. However, Andosols are also usually characterized by low inherent P availability, thus requiring higher amounts of P fertilizers than many other soils [9, 26, 27]. Extensive P fertilization on these soils may on the other hand induce high U accumulation that eventually poses risks on the environment and human health. Therefore, in this study, we aimed to evaluate high fertilization rates on Andosols with respect to fertilizer-induced U accumulation. We thus investigated U accumulation in Andosols at two long-term experimental sites in Iceland, where P fertilizers had been applied for about 50 years on permanent grasslands. Our results will provide useful information on future fertilization strategies on Andosols.

Materials and methods

Soil samples were taken from two long-term permanent grasslands in Sámstaðir and Geitasandur, Iceland, respectively (Table 1). The Sámstaðir experimental site was established in 1950 and lasted until 2004. The site is located on drained Histic Andosols, overlying a 3 m-thick Histosol, with numerous volcanic ash layers and a high input of aeolian material. For this study, we used soil samples taken from the plots that received mineral P fertilizers at an annual rate of 39.3 kg ha^{-1}

Table 1 Soil properties, fertilizer annually applied, sampling depth at Sámstaðir and Geitasandur long-term experimental sites

Treatment ID	Fertilizer annually applied (kg ha^{-1})			Soil depth	Total C	Total N	Bulk density
	N	*P*	*K*	(cm)	% DM	% DM	g cm^{-3}
Sámstaðir (1950–2004)		Histic Andosol					
0P (a)	70	0	75	0–5	15.9	1.19	0.34
				5–10	9.7	0.8	0.45
39P (e)	70	39.3	75	0–5	14.6	1.11	0.34
				5–10	8.8	0.74	0.45
Geitasandur (1958–2007)		Vitric Andosol					
0P (a1)	120	0	80	0–5	9.5	0.62	0.49
				5–10	2.2	0.17	1
39P (d1)	120	39.3	80	0–5	16.7	0.98	0.27
				5–10	2.5	0.18	0.94
80P (a2)	120	78.9[a] (55.7)	80	0–5	22.5	1.22	0.2
				5–10	3.5	0.23	0.84
80P (d2)	120	78.9[b] (67.5)	80	0–5	22.1	1.16	0.23
				5–10	3	0.2	0.89

[a] 0 kg ha^{-1} $year^{-1}$ was applied from 1958–1972; 79.6 kg ha^{-1} $year^{-1}$ was applied from 1973–2007. 55.7 kg ha^{-1} $year^{-1}$ was the average P application rate in 1958-2007

[b] 39.3 kg ha^{-1} $year^{-1}$ was applied from 1958–1972; 79.6 kg ha^{-1} $year^{-1}$ was applied from 1973–2007. 67.5 kg ha^{-1} $year^{-1}$ was the average P application rate in 1958–2007

(39P/e treatment) and plots without P fertilization (0P/a treatment). Fifteen to twenty soil cores were collected from each plot and mixed to a representative sample. Each treatment had 4 replicates.

The Geitasandur experiment started in 1958 and lasted until 2007, which was run on freely drained Vitric Andosols. The site was poorly vegetated at the start of the experiment, but a 10-cm-thick fibrous root mat was formed toward the end of the experimental period. From 1958 to 1972, a part of the site received P fertilizers at an annual rate of 39.3 (=39P/d treatment) kg ha^{-1}, while the other part received no P fertilizers (=0P/a treatment). Each treatment had three field replicates. In 1973, the original 5×10 m^2 plots were split into two 2.5 × 10 m^2 sup-plots, with one sub-plot continuing with the same P application (a1 = 0 or d1 = 39.3 kg ha^{-1} $year^{-1}$), and the other sub-plot receiving 79.6 kg P ha^{-1} $year^{-1}$ (a2 or d2 = 80P treatment). Therefore, there were three field replicates for each treatment (i.e., a1, a2, d1, d2) after 1973 (Table 1), which we used in this study. Three soil cores were collected from each plot and sub-plot.

Soil samples were taken from each of these plots at the two sites with a 20-cm-long cylindrical auger, with an inner diameter of 3.1 cm. Each soil core was further cut into 0–5, 5–10, and 10–20 cm depth intervals. This study used the soils from the depth intervals of 0–5 and 5–10 cm. More detailed information on these sites and the sampling procedures can be found in Table 1 and the studies of Gudmundsson et al. [28, 29]. A sample of superphosphate fertilizer applied at the Iceland experiment sites was collected and analyzed for its U concentration.

Soil samples were air-dried and passed through a 2 mm sieve before analysis. About 0.05 g of each soil sample was digested with a mixture of 3 ml distilled ultrapure concentrated HNO_3 (68%) and 1 ml H_2O_2 (30%, p.a.) in a pressurized microwave-assisted digestion system (UltraWave, Milestone Srl, Italy). The non-HF microwave-assisted digestion method in this study was performed according to the protocol recommended by the United States Environmental Protection Agency method 3051 [30], which has been widely applied for elemental analyses in soils. This method can extract about 80% of the total U in this study, leaving the sequestered U in structural silicate minerals as the residue [31, 32]. Three analytical replicates were carried out for each soil sample. Uranium and P concentration were determined by inductively coupled plasma mass spectrometry (ICP-MS, Agilent 7900, Germany). The analysis of the fertilizer sample was performed in the same way as the soil samples.

The differences in U concentration in various treatments were analyzed by one-way ANOVA with a significance level of $p < 0.05$. The relationships between P and U concentrations were examined by linear regression model fitting.

Annual U accumulation rates were calculated as:

$$U\ accumulation\ rate\ \left(\mu g\ kg^{-1} soil\ year^{-1}\right) = \frac{\left(U_{P-fertilized} - U_{Control}\right)}{N_{P-fertilized}}$$

where $N_{\text{P-fertilized}}$ was the number of years when P fertilizers were applied to the soil, and $U_{\text{P-fertilized}}$ and U_{Control} were U concentrations in the soils with and without P fertilizers, respectively.

Fertilizer-derived U accumulation

The U concentration in one of the applied P fertilizers at the two sites was 95 mg U kg^{-1} fertilizer. After application for about 50 years of such a fertilizer at a rate of 39.3 kg P ha^{-1}, soil U concentrations in the surface soil (0–5 cm) increased by 7.3 mg kg^{-1} and 6.1 mg kg^{-1} at Geitasandur and Sámstaðir, respectively. Moreover, the U concentrations in the surface soil of the 80P treatments exceeded 15 mg kg^{-1} and were almost twice that of the 39P treatments in Geitasandur (Fig. 1a).

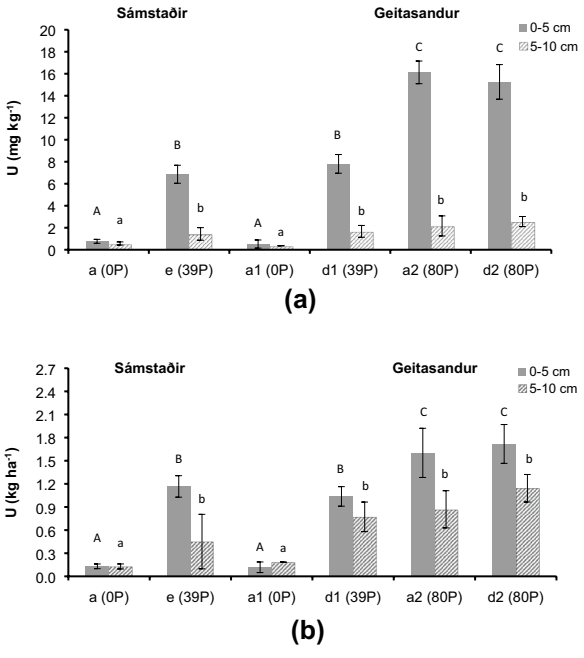

Fig. 1 Uranium concentrations (**a**) and stocks (**b**) in Andosol soils with the treatments of different P application rates at Sámstaðir and Geitasandur long-term experimental sites in Iceland. Error bars are the standard deviation of treatment replicates. Different letters indicate significant differences among the treatments with a significant level of $p < 0.05$

No significant difference was found in U concentrations between 0–5 cm and 5–10 cm soil depth in the controls at both sites. However, in the 39P and 80P treatments, U concentrations increased not only in the 0–5 cm but also in the 5–10 cm soil depth, increasing by 1 and 2 mg kg^{-1} for 39P and 80P treatments, respectively (Fig. 1a).

The U stocks in the top 0–5 cm soil were significantly smaller in the control than in the P fertilization treatments, so were the U stocks in the 5–10 cm depth (Fig. 1b). In the control, there was no significant difference in U stock between 0–5 cm and 5–10 cm soil, whereas this difference was significant in the P fertilizer treatments both at Sámstaðir and Geitasandur (Fig. 1b). Compared with the control, 1.04 kg U ha^{-1} had been added at Sámstaðir (0–5 cm) under a P application rate of 39.3 kg ha^{-1}, while 0.92 and 1.55 kg U ha^{-1} had been added at Geitasandur (0–5 cm) in the 39P and 80P treatments, respectively, over a period of about 50 years (Fig. 1b). In the soil depth of 0–5 cm, the final fertilizer-derived U input was about ten times that in the control. In the course of the experiments at both sites, over 60% of fertilizer-derived U had accumulated in the top 0–5 cm soil (Table 2).

The annual U accumulation rates (0–5 cm) were 113, 149, 310 μg kg^{-1} $year^{-1}$ for the 39P (e), 39P (a1) and 80P (a2, d2) treatments, respectively. In addition, the amounts of annual U input to one hectare were 19, 19 and 32 g ha^{-1} $year^{-1}$ (Table 2).

The concentrations of P and U correlated significantly in both soils ($R^2 > 0.7$, $P < 0.05$; Fig. 2), confirming that the U accumulation in the grassland most likely coincided with P accumulation under the P fertilizer applications.

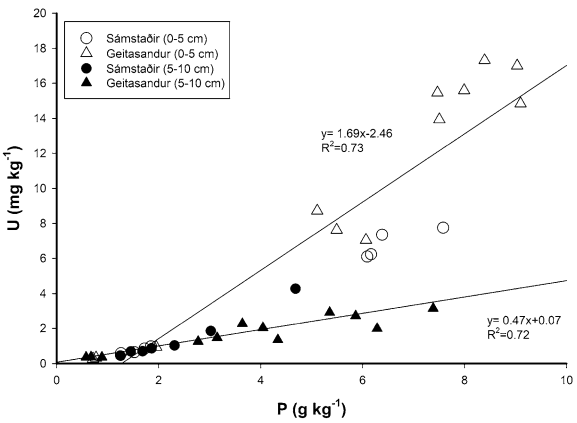

Fig. 2 Relationships between P and U concentrations in soil depth of 0–5 cm and 5–10 cm in Sámstaðir and Geitasandur

Discussion

Our results support the earlier findings that U accumulates in agricultural soils due to P fertilization [5–7, 9]. However, the annual U accumulation rates found in this study exceeded those reported for other ecosystems so far. The U accumulation rates in the top 5 cm reported in this study (113, 149 μg kg^{-1} $year^{-1}$) were far above the high end of the to-date reported U accumulation rates (2–29 μg kg^{-1} $year^{-1}$) for other soils with similar P application rates (30–45 kg ha^{-1} $year^{-1}$) [7, 11, 33]. Even though a broad range of U accumulation rates (0–130.6 μg kg^{-1} $year^{-1}$) were found in previous studies [5–11, 16, 24, 33–35], none of them reached the values found in the present study (Table 2).

Clearly, the amount of the applied P fertilizers is one of the critical factors for the amount of U that accumulates in soils. In our study, U accumulation rates increased with increasing amounts of P fertilization. The P application

Table 2 Uranium accumulation rates and U inputs at Sámstaðir and Geitasandur long-term experiment sites

Soil depth	Sámstaðir	Geitasandur		
	39P (e)	39P (a1)	80P (a2)	80P (d2)
0–5 cm				
Annual accumulation rate (μg kg^{-1} $year^{-1}$)	113	149	319	301
U input (g $ha^{-1}year^{-1}$)	19	19	30.3	33.2
U in applied P fertilizers (mg U kg^{-1} P)	489	482	545	492
5–10 cm				
Annual accumulation rate (μg kg^{-1} $year^{-1}$)	16	27	37	45
U input (g $ha^{-1}year^{-1}$)	4	12	15	20
0–10 cm				
Annual accumulation rate (μg kg^{-1} $year^{-1}$)	58	52	87	94
U input (g $ha^{-1}year^{-1}$)	23	31	45	53
U in applied P fertilizers (mg U kg^{-1} P)	580	794	811	782

Table 3 To-date reported U accumulation rates in Andosols

Country	Experiment site (Andosols)	P application rate (kg ha^{-1} year^{-1})	U accumulation rate (μg kg^{-1} year^{-1})	Reference
Japan	Fujisaka Branch	61	4.2	Takeda et al. [5]
	–	74.3	130.6	Yamaguchi et al. [9]
New Zealand	Taupo	19.7	15	Taylor [6]
	Hinemaiai	98	47	Taylor [6]
	Whatawhata	30	0	Schipper et al. [7]
		50	42	
		100	67	

rate (79 kg P ha^{-1} yr^{-1}) at Geitasandur was twice or three times that typically applied to non-Andosols, thus leading to the higher U accumulation rates. A high U accumulation rate of 130.6 μg kg^{-1} year^{-1} was also found in an Andosol in a long-term experiment in Japan with an annual P application of 74.3 kg ha^{-1} year^{-1} [9] (Table 3). Accumulation rates of U reported in Andosols of New Zealand were 15–67 μg kg^{-1} year^{-1} with a P application range of 19.7–100 kg ha^{-1} year^{-1} [6, 7] (Table 3). Risks of fertilizer-derived U should thus be specifically considered on Andosols.

Uranium concentration in the applied P fertilizer is another factor determining the U accumulation rate [5, 36]. When U concentration in the applied P fertilizer is low, low U accumulation rate can also occur in Andosols. Takeda et al. (2006) found a relatively low U accumulation rate of 4.2 μg U kg^{-1} year^{-1} in an Andosol in Japan in spite of a high P application of 65 kg ha^{-1} year^{-1} [5] (Table 3). This was attributed to the low U concentration (31 mg U kg^{-1} fertilizer) in the applied superphosphate [5]. The differences in U concentrations of P fertilizers are attributed to the variability of U concentrations in different phosphate rocks used for P fertilizer production. In general, igneous phosphate rocks (e.g., from Russia) usually contain less U (2.5–40 mg kg^{-1}, mean value 14.4 mg kg^{-1}) than sedimentary rocks (e.g., from Morocco) (57–245 mg kg^{-1}) [1, 36]. In addition, U concentrations in sedimentary phosphate rocks differ in various deposition environments [1, 3, 36]. The phosphate rocks imported to Europe are predominately from Morocco (35.1%), Russia (31.6%), Algeria (12.3%) and Israel (7.5%) [37]. As these phosphate rocks are either igneous or sedimentary, their U concentrations would also vary in a wide range. Therefore, for soils like Andosols which require large amounts of P fertilizers, selecting fertilizer products low in U should be a sustainable way to both ensure crop yields and minimize fertilizer-derived U accumulation.

To evaluate the P fertilizer quality regarding U concentration, we analyzed a P fertilizer sample applied at the two sites. Its U concentration (95 mg U kg^{-1} fertilizer) was at the middle level in the range of previously reported values (21–272 mg kg^{-1} fertilizer) [38]. However, as the fertilizers applied during the 50 years also included other superphosphates with unknown U concentrations, we in addition estimated an average U concentration per kg P using the total increased U stock divided by the total amounts of the applied P, which resulted in 580 and 795 mg U kg^{-1} P for Sámastaðir and Geitasandur, respectively. Since superphosphates contain about 8.7% P, the U concentration per kg fertilizer was then 50.5 and 69.2 mg U kg^{-1} fertilizer, respectively. Again, these values were within the range of the reported U concentrations in P fertilizers. Nevertheless, the application of these fertilizers that contained U in a normal range resulted in a remarkable increase of soil U concentrations at our study sites.

It is worth noting that the average annual U inputs were 19 and 32 g ha^{-1} year^{-1} when P was applied at a rate of 39.3 and 78.9 kg ha^{-1} year^{-1}, respectively. These values were within the range of previously reported values (8.6–47 kg ha^{-1} year^{-1}) [5, 39, 40]. Our soils thus exhibited a characteristic U accumulation with high accumulation rates but meanwhile with the moderate annual U inputs. We attribute this observation to the low bulk densities of our Andosols (0.2–0.5 g cm^{-3} in the 0–5 cm soils). Compared with non-Andosols, such low bulk densities resulted in a lower total weight of the soil within a given area and depth, thus increasing the U concentrations and the U accumulation rates. In addition, Andosols usually exhibit elevated contents of organic matter [27], which promote the retention of U in the very surface soil [23]. Besides, no tillage for such a long period of fertilization also contributed the high U concentration in the top 5 cm of those two fields.

Conclusions

In this study, we report two cases of high fertilizer-derived U accumulations at the long-term experiment sites in Iceland (Sámstaðir and Geitasandur). This

resulted from a combined effect of two main factors. First, large amounts of P fertilizers were applied to these Icelandic Andosols to maintain grassland productivity because of inherent low P availability in Andosols. Second, the low bulk density and high organic matter content in Andosols effectively concentrated U in the upper surface soil. These two factors also play a role in agriculture systems other than Andosols, e.g., on former peatlands, raising the possibility that more unreported areas of agricultural land could contain U with a concentration close to or even higher than the (sole) soil quality guideline of 23 mg U kg^{-1}. Therefore, for these types and other agricultural ecosystems requiring high amounts of P fertilization, proper selection of those P fertilizers low in U content will therefore be particularly important for sustainable land use.

Abbreviations
P: Phosphorus; U: Uranium; ICP-MS: Inductively coupled plasma mass spectrometry.

Acknowledgements
The first author would like to thank the supported from the Chinese Scholarship Council (CSC201506350148) for her study in Germany.

Authors' contributions
TG collected soil samples. YS analyzed the samples. YS, WA, TG and RB did the data interpretation. YS was the major contributor in writing, with WA, BW, TG and RB contributed to the writing. All authors read and approved the final manuscript.

Funding
Chinese Scholarship Council (CSC201506350148).

Availability of data and materials
All data analyzed during this study are available from the corresponding author on reasonable request.

Ethics approval and consent to participate
Not applicable.

Consent for publication
Not applicable.

Competing interests
The authors declare that they have no competing interests.

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Received: 1 March 2020 Accepted: 15 June 2020
Published online: 01 July 2020

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